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Modelling critical behaviour in terms of catastrophe theory and fractal lattices

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Abstract. A D -dimensional lattice model for a one-component fluid system exhibiting critical behaviour is set up. The free energy per unit volume of the lattice can be described by Thom's cusp singularity if $D = 4 - \varepsilon$ and $\varepsilon \approx 0$. The model indicates that a non-diffeomorphic relationship between the parameters of topological singularity theory and measurable quantities is feasible and leads to critical exponents which agree quite well with the observed ones. The non-integral dimension D of the lattice is interpreted in terms of fractals.

1. Introduction

The possibility of using Thom's catastrophe theory to study phase transitions has been suggested during the last few years (e.g. Schulman 1971, 1973, Thom 1972, 1975, Schulman and Revzen 1972, Fowler 1972, Benguigui and Schulman 1973, Gilmore 1977, de Alfaro and Rasetti 1978, Poston and Stewart 1978, Grandy 1979, Dukek 1979, Keller *et al* 1979, Vendrik 1979). Topological features of phase diagrams near isolated n th-order phase transition points ($2 \leq n \leq 4$) have been predicted in a qualitatively excellent way. The principal advantage of applying catastrophe theory to critical phenomena is that it classifies the possible singularities of thermodynamic potentials and gives the free energy directly in polynomial form, so that no arbitrary truncation of a power series expansion of the free energy is required as in the Landau theory (e.g. Landau and Lifschitz 1975).

Near higher-order phase transition points, the dependence of the relevant physical quantities on the reduced temperature $t = (T - T_c)/T_c$ is given—in first approximation—by power laws. T_c will denote the value of the temperature T at the n th-order phase transition point. The exponents of these power laws are the n th-order critical exponents (for a detailed definition see e.g. Kadanoff (1970), Stanley (1971), Ma (1976)). The objective of this paper is to calculate from catastrophe theory critical exponents ($n = 2$) which agree with experimentally measured critical exponents.

Let us give a short review of the work done on critical exponents in catastrophe theory. Up to now, there have been two approaches for the application of catastrophe theory to phase transition phenomena and critical exponents.

The first one starts from some special thermodynamic equation of state near the critical point of interest. The most famous example is the van der Waals equation (see e.g. Fowler (1972), Poston and Stewart (1978)). The physical reduced variables are

transformed to the coexistence set of some normal form (the cusp normal form for the example of the van der Waals equation). This transformation turns out to be diffeomorphic, i.e. reversible and infinitely often differentiable, at least for the special cases usually analysed. If the normal form is identified, and if the (diffeomorphic) relation between parameters of catastrophe theory and reduced physical variables is known, then it is possible to calculate critical exponents. This method reproduces the critical exponents of Landau theory (e.g. Poston and Stewart 1978).

If one attempts to improve the numerical values for the critical exponents, a suggestive first trial is given by the change of Thom's 'Maxwell convention', because the coexistence set depends on the convention chosen to decide between competing minima of the normal form. But it has been proved that no change of the convention will alter the exponents (Poston and Stewart 1978).

However, we want to emphasise that this method only describes a small subset of thermodynamic state equations by means of catastrophe theory. There is, for example, no diffeomorphic transformation known from Schofield's equation of state (e.g. Schofield 1969, Kortman 1972) to a coexistence set of some normal form.

More powerful is the second approach (e.g. Schulman 1971, Schulman and Revzen 1972, Vendrik 1979). It starts from some thermodynamic potential $G: X \times U \rightarrow \mathbb{R}$, with $G_u(\mathbf{x})$ bounded below, and assumes $G(\mathbf{x}, \mathbf{u}_0)$ to be a 'singularity' at some point $\mathbf{u}_0 \in U$. X is the thermodynamic state space and U some space of external 'control' variables. ' $G(\mathbf{u}, \mathbf{x})$ a singularity' means, more precisely, that the family $G_u(\mathbf{x})$ has a singular member $G_{\mathbf{u}_0}(\mathbf{x})$, dividing $G_u(\mathbf{x})$ into qualitatively different classes. If $G(\mathbf{u}, \mathbf{x})$ is a smooth function with respect to \mathbf{x} and $\dim U \leq 14$ (Arnold 1974), then the structurally stable unfoldings of that singularity can be classified with the help of the normal forms of catastrophe theory. That means almost all structurally stable unfoldings of $G(\mathbf{x}, \mathbf{u}_0)$ are diffeomorphic to some normal form specific for the order of the singularity $G(\mathbf{x}, \mathbf{u}_0) = G_{\mathbf{u}_0}(\mathbf{x})$.

If one assumes (\mathbf{x}, \mathbf{u}) to be analytic functions of some reduced physical variables, the above approach reproduces the Landau theory and the critical exponents of Landau theory (Schulman 1973, Poston and Stewart 1978, Keller 1979). Therefore non-analytic relations between physical variables and catastrophic variables have been assumed—but on an entirely *ad hoc* basis (Benguigui and Schulman 1973, Schulman 1973).

In this paper we set up a model that justifies a non-analytic relation. The idea of our model originates from the simple statement that critical exponents calculated in a two-dimensional Ising model are quite different from the critical exponents calculated in a three-dimensional Ising model, although in both models the type of the Hamiltonian is the same. Obviously the temperature dependence of the magnetisation is related to the dimension of the Ising model.

In § 2 we propose a model for a one-component fluid. The model is a straightforward generalisation of the lattice-gas model. Imagine that for each of N different samples of the fluid m measurements have been made at time t_0 . The result is a set of N vectors $\mathbf{w}_i \in \mathbb{R}^m$ ($1 \leq i \leq N$). To introduce an arbitrary spatial order on the above set of measurements, we assign each vector to one and only one point \mathbf{z}_i of a lattice L with dimension D . If N measurements completely describe the real fluid for a given time t_0 , then the lattice L is a complete and static description of the fluid for the time t_0 . Now the particles of the fluid interact mutually and, therefore, repeating the set of measurements at the same parts of the fluid, but at some time $t > t_0$, will most probably not reproduce the same results; i.e. $\mathbf{w}_i(t) \neq \mathbf{w}_i(t_0)$. To simulate this effect, one has to assume an

interaction between the vectors fixed at each lattice point. We introduce a Hamiltonian confined to nearest-neighbour interaction.

For the example of the one-component fluid, we specify the measurement vectors to have only one component, i.e. to be scalars (the 'occupancy state variable'). This specification is not crucial and is only made for simplification.

The next step is to define unit volumes with the lateral dimension of the order of the correlation length ξ . We introduce a free energy F per unit volume and observe the effect on F if the correlation length is enlarged. We show that near an isolated second-order phase transition point the free energy per unit volume is represented by the unfolding of the cusp singularity $V_2(x, 0, 0)$.

In § 3, we attack the determination of the dimension D of the lattice via scaling arguments applied to the cusp normal form and to the correlation function. It is found that D depends on the codimension of the cusp normal form and some small parameter η ($0 \leq \eta \leq 0.1$). In accordance with renormalisation group considerations, the dimension of the abstract lattice will be approximately 4 ($3.8 \leq D \leq 4$). Somewhat arbitrarily, we choose $D = 3.92$ and again use scaling arguments, this time applied to the abstract lattice ($D = 3.92$) and to the real fluid ($D = 3$). This method gives relations between:

(1) the abstract mathematical order parameter x and the reduced physical density ρ ;

(2) the unfolding parameter a_2 and the reduced temperature t .

We insert the relations $x(\rho)$ and $a_2(t)$ in the standard cusp normal form and calculate critical exponents. These exponents agree quite well with the experimental observations for CO_2 , Xe and He^3 (Stanley 1971, Ma 1976).

The 'broken' dimension $D = 3.92$ is a potential embarrassment to the human mind. For this reason the mathematical concept of spaces with non-integral dimension is introduced in § 4; however, in a more speculative way. We illustrate an example for a space with non-integral Hausdorff-Besicovitch dimension (Mandelbrot 1977) and define 'fractal lattices' (see also Gefen *et al* (1980)). Scaling, differentiation and integration remain unchanged on fractals, and therefore our arguments leading to the normal form V_2 and the relations $x(\rho)$, $a_2(t)$ are not affected if the dimension of L is broken. However, no attempt is made to elucidate the physical meaning of the fractal lattice L , i.e. to relate clustering in the fractal lattice or the real fluid to the dimension of fractal lattice and fluid.

In the last section we summarise our findings and outline developments for higher-order phase transition phenomena (neglecting logarithmic corrections).

The Appendix will give some additional information for readers not familiar with the application of catastrophe theory to phase transition phenomena.

2. A lattice model of a fluid system

We consider a one-component fluid system made up of N molecules ($N = 10^{23}$). If v is roughly the volume of one molecule (typically a few \AA^3), we divide the whole volume V of the fluid system into N_0 cells of size v , $N_0 = V/v$. For a fluid system we surely obtain $N_0 > N$. Now we introduce the occupancy e as a state variable. A cell is in the occupied state $e = 1$ if it is occupied by the centre of one molecule. Otherwise the cell is said to be in the empty state $e = -1$ (Kittel 1969, Stanley 1971).

An abstract lattice model of the fluid is constructed as follows. We measure the occupancy e for each cell to obtain a complete set of N_0 measurements e_1, e_2, \dots, e_{N_0} .

Then each measured value e_j is fixed to one and only one lattice point z_j of the N_0 points of a D -dimensional lattice L . By this, each cell (or subsystem) corresponds exactly to one site z of a D -dimensional isotropic lattice L , with lattice spacing Λ^{-1} . At each lattice site, the micro-observable $e(z)$ (i.e. the state of the corresponding subsystem) has the value 1 or -1 . Neighbouring cells do not necessarily correspond to neighbouring lattice sites, because for $D \neq 3$ such a correspondence is quite impossible to establish.

We further replace the real interaction in the fluid system by an artificial one in the lattice model, analogous to the familiar lattice-gas model (Kittel 1969, Stanley 1971). This means we assume that interaction takes place only between neighbouring lattice sites. Obviously, $D > 3$ corresponds to something like averaged long-range interaction in the real fluid because each cell interacts with more than six other cells.

The partition function Z of the lattice is given (Stanley 1971) by

$$Z = \sum_{e(z)}^* \exp\left(-\beta J \sum_{\langle\langle zz' \rangle\rangle} e(z)e(z')\right) \quad (2.1)$$

where $\beta = 1/kT$, k is the Boltzmann constant, T is the temperature, J is a coupling factor and $\langle\langle zz' \rangle\rangle$ denotes summation over nearest neighbours z' . The asterisk in (2.1) means that the summation in (2.1) is restricted to those configurations which satisfy

$$\sum_{z \in L} e(z) = N. \quad (2.2)$$

The total free energy F is given by

$$F = -\beta^{-1} \ln(Z). \quad (2.3)$$

For the order parameter y (the ensemble average of e) we obtain

$$y = \langle e \rangle = \frac{1}{Z} \sum_{e(z)}^* e(z) \exp\left(-\beta J \sum_{\langle\langle zz' \rangle\rangle} e(z)e(z')\right). \quad (2.4)$$

The following technical assumptions will be made.

(A1) The lattice is infinitely extended. This is reasonable because N is very large.

(A2) For calculations we can replace the sums in (2.1)–(2.4) by integrals.

The first assumption implies that the partition sum and the total free energy become infinite. To obtain finite quantities, the theorem of the thermodynamic limit is invoked so that it is possible to divide the infinite lattice into finite sublattices or blocks, which are essentially similar to the whole lattice. These blocks will be called 'unit volumes', and the extension of each block is large in comparison with Λ^{-1} , but the extension is in the order of magnitude of the correlation length. Then (2.1) can be replaced by the approximation

$$Z \approx \prod_{i=1}^{\infty} Z_i \quad (2.5)$$

where Z_i is the partition sum of one unit volume. From (2.5) it follows that

$$F \approx \sum_{i=1}^{\infty} F_i \quad (2.6)$$

where F_i is the free energy per unit volume i , $F_i = -\beta^{-1} \ln(Z_i)$. Since all blocks are assumed to have the same size, we have $F_i \approx F_j$ for all i, j and so can drop the subscripts, i.e. define $F = F_i =$ 'free energy per unit volume'. Disregarding small statistical fluctuations, the order parameter y has the same value in the unit volume and in the whole

lattice. If one knows the behaviour of the order parameter in one block, one knows the behaviour of the order parameter in the entire system.

One block in the lattice corresponds to an ensemble of cells in the real fluid. Therefore the order parameter y should be related to the fluid density $\tilde{\rho}$.

Now the question arises of how large a block should be if one changes the reduced temperature t . Since the correlation length ξ diverges for $T \rightarrow T_c$, the renormalisation group procedure must be applied to the abstract lattice (Ma 1973, 1976, Wilson and Kogut 1974). This means that we have to collect lattice sites inside a volume s^D , then calculate the mean value \bar{e} of $e(\mathbf{z})$ in s^D and then assign this mean value to a site \mathbf{z}' of a lattice of the same kind, i.e. with the same dimension, extension, lattice spacing and unit volumes. Therefore the renormalisation group can be interpreted as a symmetry group \hat{R} on the abstract lattice (Ma 1976). One element $R_s \in \hat{R}$ changes $e(\mathbf{z})$ continuously in s to a new value $e'(\mathbf{z}) \in [-1, 1]$, thus smoothing $\{e(\mathbf{z})\}$. Furthermore, R_s generates powers of $e(\mathbf{z})$ in the Hamiltonian of the transformed partition function. The calculation of $H' = R_s H$ requires assumptions (A1) and (A2).

We need only two results of renormalisation group theory. The first one states that the correlation length scales as $\xi \propto |t|^{-\nu}$ and $\nu = \frac{1}{2}[1 + (4-D)/6 + \dots]$ for critical phenomena (Wilson and Kogut 1974). This will be used in § 3. The second states that in each case $-1 \leq e(\mathbf{z}) \leq 1$. Then the ensemble average $y = \langle e \rangle$ is limited too, i.e. $-1 \leq y \leq 1$.

From (2.1), (2.3) and (2.4), it is obvious that the free energy is a continuous function of the order parameter y even after the application of R_s . Likewise $F(y)$, the free energy per unit volume, is continuous on the compact interval $[-1, 1] \in \mathbb{R}$ and for that reason—due to Weierstrass— $F(y)$ can be approximated by a series expansion

$$\tilde{F}(\mathbf{u}, y) = \sum_{j=1}^M u_j y^j$$

and $|\tilde{F} - F| \rightarrow 0$ if $M \rightarrow \infty$. Replacing the sums in (2.1)–(2.4) by integrals, we obtain

$$\prod_{\text{all } \tilde{F}} \int dy \exp[-\beta \tilde{F}(y, \mathbf{u})] \approx \int d\mathbf{e}(\mathbf{z}) \exp\left(-\beta \int_L d^D \mathbf{z} H(\mathbf{e}(\mathbf{z}), \mathbf{g})\right). \quad (2.7)$$

Here \mathbf{g} denotes a multidimensional parameter generated by \hat{R} from the small number of parameters in the initial Hamiltonian (Ma 1976, Wilson and Kogut 1974).

From (2.7) and (2.1) it follows that minima of \tilde{F} correspond to maxima of Z and, therefore, to macroscopic states (see e.g. Kittel (1969)). Hence, the set

$$\Sigma = \{(y, \mathbf{u}) | \partial \tilde{F}(y, \mathbf{u}) / \partial y = 0 \text{ and } \partial^2 \tilde{F}(y, \mathbf{u}) / \partial y^2 \geq 0\}$$

describes macroscopic states (distinguished by the order parameter). At a critical point $\mathbf{u} = \mathbf{u}_0$ in the phase diagram two macroscopic states merge into only one macroscopic state. This merging (or branching) of states implies that $\tilde{F}(y, \mathbf{u}_0)$ possesses powers in y higher than third order, i.e. $\tilde{F}(y, \mathbf{u}_0) = u_{04} y^4 + u_{05} y^5 + \dots$. For this reason, criticality implies that $\tilde{F}_0 = \tilde{F}(y, \mathbf{u}_0)$ is a singularity not of Morse type but of Thom type. Let \mathbf{u}_0 be the origin in $U = \{\mathbf{u} | u_i \in \mathbb{R}\}$ and $\tilde{y} = (y - y_0) / y_0$, where $y_0 = y(\mathbf{u}_0)$ is the value of y at the critical point \mathbf{u}_0 ; then $\tilde{F}_0 = \tilde{F}(\tilde{y}, 0)$.

In (2.7) \tilde{F} must be assumed to be bounded below—otherwise the integral does not exist. In this case, according to Thom's theorem, the simplest singularity \tilde{F}_0 has codimension two and its stable unfoldings are diffeomorphic to the cusp normal form V_2 (Thom 1975). This means that there exists a diffeomorphism ψ which transforms (\tilde{y}, \mathbf{u})

into $(x, a_1, a_2, a_3, \dots)$. Only the parameters x, a_1, a_2 are relevant because the remaining parameters a_3, \dots merely give rise to a remainder $Q(a_3, \dots)$ independent of x . Q will not affect critical behaviour. Even if \tilde{y} is not a one-dimensional state variable (as in the fluid case) but an r -dimensional state variable, the statement remains true that Q does not affect critical behaviour; then the remainder Q is a quadratic term of Morse type in at least $r-2$ state variables. Consequently, up to diffeomorphisms and an irrelevant term Q , the free energy F^* per unit volume is given by the normal form of the cusp unfolding, and F^* is no truncation, but an exact polynomial:

$$F = \underset{C^\infty}{\tilde{F}}(\tilde{y}, \mathbf{u}) \propto F^*(x, \mathbf{a}) = \frac{1}{4}x^4 + \frac{1}{2}a_2x^2 + a_1x + Q. \quad (2.8)$$

The symbol ' $A \underset{C^\infty}{\propto} B$ ' means that A is diffeomorphic to B . For fixed a_1, a_2 the minima of F^* give the stable states, i.e. the deepest minima of V_2 give the most probable stable states. That is the reason for the so-called 'Maxwell convention'. This convention is due to Thom, and states that the deepest minimum of a normal form gives the physical stable state.

In what follows, we are interested in the relation Φ between x, a_1, a_2 and measurable quantities of the fluid such as the reduced density $\rho = (\tilde{\rho} - \tilde{\rho}_c)/\tilde{\rho}_c$, the reduced temperature $t = (T - T_c)/T_c$, etc. Gibbs' phase rule requires there to be precisely two independent variables in the phase diagram of the fluid. We choose for them t and the reduced pressure $p = (P - P_c)/P_c$. Consequently, the relation $(x, a_1, a_2) = \Phi(\rho, p, t)$ is bijective.

3. Scaling and critical exponents

As the fluid system approaches the critical temperature T_c , the correlation length ξ diverges as $\xi \propto |t|^{-\nu}$ if $t \rightarrow 0_+$ and $\xi \propto |t|^{-\nu'}$ if $t \rightarrow 0_-$. Therefore we have to enlarge our unit volume by the factor ξ , as sketched in figure 1, where the effect of one element R_s on the lattice is illustrated. Let us now assume that the fixed point of \hat{R} in parameter space is reached, and let F_0^* be the free energy per unit volume at this fixed point. To find the dependence of F^* on x, a_1, a_2 , we have to move away slightly from the fixed point (critical point), i.e. to unfold F_0^* . This means in principle, that we have to 'invert' the renormalisation procedure, and apply R_s^{-1} to H_0^* , where H_0^* is the Hamiltonian giving F_0^* . This cannot be done explicitly, because it is impossible to construct one definite member of an ensemble from some mean values of that ensemble. However, scaling arguments remain unaffected.

R_1 is the identity in \hat{R} , and from

$$R_s R_t = R_{st} \quad (3.1)$$

one concludes that $R_s^{-1} = R_{s^{-1}}$. It is easily seen that the correlation length shrinks if we move away from the fixed point, and $R_s^{-1} = R_{s^{-1}}$ implies that we have to reverse the sign in the exponent of $\xi(t)$. 'Inverting' the renormalisation procedure means, therefore, length scaling by $s \propto |t|^\nu$ if $t > 0$ and by $s \propto |t|^{\nu'}$ if $t < 0$.

A given volume V_1 transforms according to

$$V_s = s^{-D} V_1. \quad (3.2)$$

The total free energy naturally is invariant under \hat{R} . Then, according to (3.2), the free

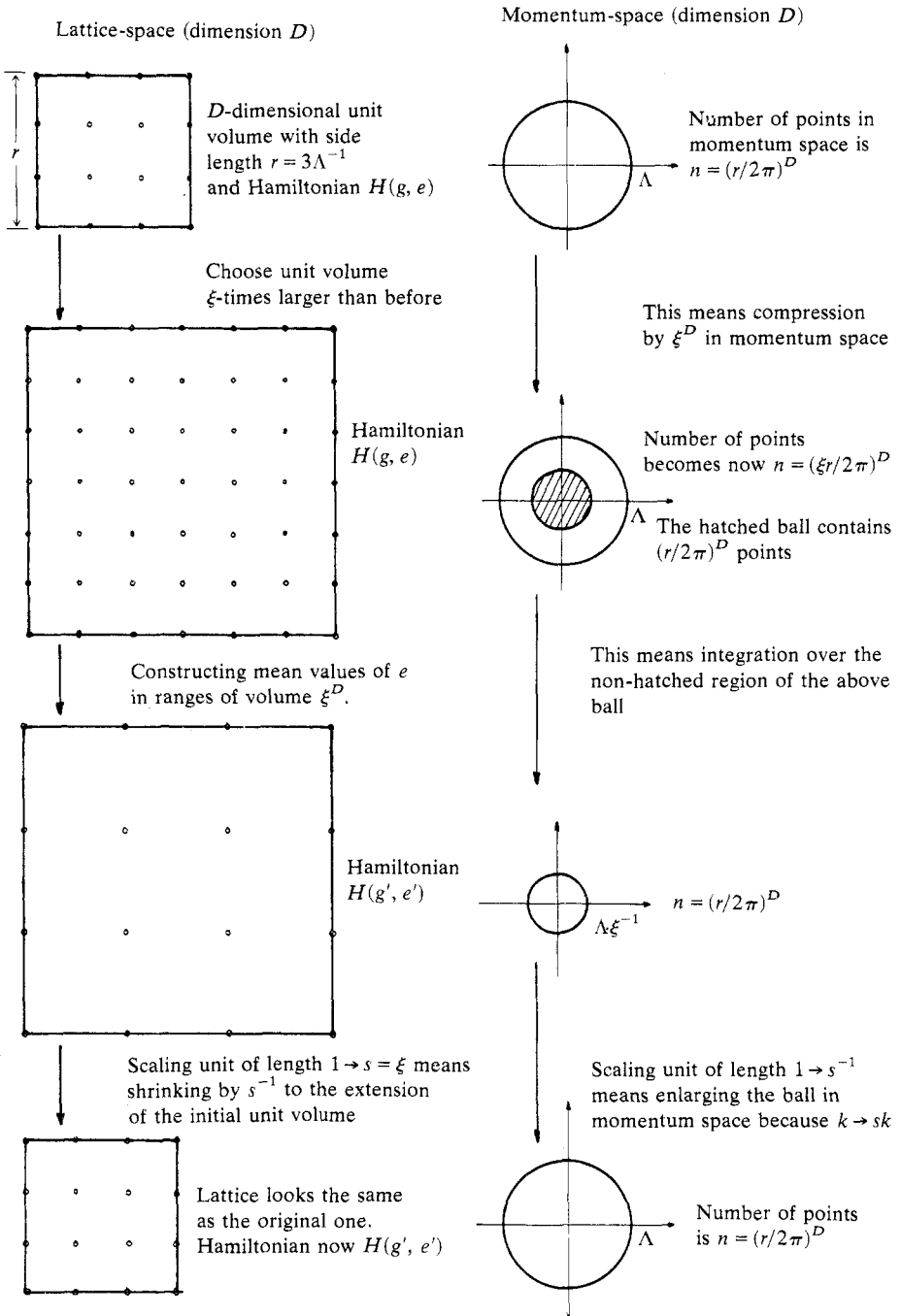


Figure 1. Application of R_s with $s = 2$ to a unit volume.

energy per unit volume transforms as

$$F_s = s^D F_1. \quad (3.3)$$

If the lattice model is to be a reasonable approximation to the fluid, then a change of the correlation length in the fluid and in the lattice should produce the same effect in the free energies per unit volume of both systems. We change the length scale in the lattice by s and in the fluid by r according to

$$r \propto |t|^{\nu_2}, \quad (3.4a)$$

$$s \propto |t|^{\nu_1}. \quad (3.4b)$$

The requirement that a change of the correlation length produces the same effect in both the fluid and the lattice means equality of the free energies per unit volume in both systems after the transformation, if these free energies were equal before in the fluid (subscript 2) and in the lattice (subscript 1), i.e. $r^{D_2} \propto s^{D_1}$. From (3.3) and (3.4) it then follows that

$$D_1 \nu_1 = D_2 \nu_2 = 3 \nu_2. \quad (3.5)$$

We now use (3.3), (3.5) and the correlation function $G(k)$ in the lattice L to determine $x(\rho)$. Let $e^* = (e - e_c)/e_c$ be the reduced micro-order parameter, where e_c is the value of e at the critical point. The correlation function is defined (e.g. Ma 1976) by (neglecting the subscript '1')

$$G(\mathbf{k}) = \int_V d^D \mathbf{z} \langle (e^*(\mathbf{z}) - \langle e^* \rangle)(e^*(\mathbf{0}) - \langle e^* \rangle) \rangle e^{-i\mathbf{k}\mathbf{z}}. \quad (3.6)$$

If the volume V of integration is large enough, (3.6) can be rewritten as

$$G(\mathbf{k}) = \frac{1}{V} \int d^D \mathbf{z} d^D \mathbf{z}' \langle e^*(\mathbf{z}) e^*(\mathbf{z}') \rangle e^{-i\mathbf{k}(\mathbf{z}-\mathbf{z}')}. \quad (3.7)$$

From this it follows that $G(\mathbf{k}) \propto (e^* \text{ density})^2 \times (\text{volume})$. A change of the unit of length yields $\langle e^* \rangle \rightarrow s^q \langle e^* \rangle$, whence

$$G(s\mathbf{k}) \propto s^{2q-D}. \quad (3.8)$$

On the other hand, it is known (Stanley 1971, Ma 1973, 1976) that

$$G(\mathbf{k}) \propto |\mathbf{k}|^{-2+\eta} \quad (3.9)$$

and this scales as

$$G(s\mathbf{k}) \propto s^{-2+\eta}. \quad (3.10)$$

Comparing (3.8) with (3.10), we obtain

$$q = (D - 2 + \eta)/2. \quad (3.11)$$

On the coexistence line one has $a_1 \equiv 0$ and $a_2 = -x^2$ from (2.8) and the Maxwell convention. Therefore $V_2(x, a_1, a_2) = -\frac{1}{4}x^4$ on the coexistence line, and from (3.3) it follows that $x^4 \propto s^D$ or

$$x \propto s^{D/4}. \quad (3.12)$$

Now, due to (2.8), $x \propto_{C^\infty} \hat{y} = \langle e^* \rangle$ and comparing (3.11) with (3.12), it is seen that for the cusp normal form

$$\frac{1}{2}D = 2 - \eta \tag{3.13}$$

or

$$\eta = 2 - D/2.$$

We note that $\eta = 0$ for $D = 4$; i.e. $x \propto s^q$ with $q = 1$.

The experimental value for the exponent η is very small, namely $0 \leq \eta \leq 0.1$. That value inserted in (3.13) limits the dimension to the range $3.8 \leq D \leq 4$. In other words, the cusp model gives a good approximation to the free energy per unit volume only if the dimension of the abstract lattice is close to four. We choose $D = 3.92$, because this produces a good numerical fit for the critical exponents.

One can calculate the correlation function for the real fluid ($D = 3$), and the same arguments as in (3.6)–(3.11) lead to

$$q_2 = (3 - 2 + \eta_2)/2 = (1 + \eta_2)/2. \tag{3.11a}$$

From the change of the reduced density ρ of the fluid ($D = 3$) with t we obtain, using (3.11a), the relation $\rho(x)$:

$$\rho \propto r^{(1+\eta_2)/2} \propto t^{\nu_2(1+\eta_2)/2} \propto (t^{\nu_1})^{D_1(1+\eta_2)/6} \propto (x^2)^{(1+\eta_2)/3}$$

or equivalently, with $\theta = 3/[2(1 + \eta_2)]$,

$$x(\rho) \propto \rho^\theta. \tag{3.14}$$

The results obtained so far do not depend on the sign of the exponent in (3.4). However, to determine the form of $a_2(t)$ this sign plays an important role. On the coexistence line we have $a_2 = -x^2$ as stated above. From the scaling of x^2 by s it follows for a_2 that

$$a_2(t) \propto t^\lambda \tag{3.15}$$

with $\lambda = \nu_1 D_1/2$.

Renormalisation group considerations (Wilson and Kogut 1974) show that $2\nu = 1 + \varepsilon/6 + \varepsilon^2/25 \pm \dots$ and $\varepsilon = 0.08$ gives $\nu_1 = 0.506$. From (3.4) we find for the fluid that $\nu_2 = \nu_1 D_1/3 = 0.662$ and (3.12) yields $\eta_1 = \varepsilon/2 = 0.04$.

The value of η_2 cannot be deduced from the model: we assume $\eta_2 = 0.08$. Then equations (3.14) and (3.15) give the exponents $\theta = 1.39$ and $\lambda = 0.99$, i.e. $x \propto \rho^{1.39}$, $a_2 \propto t^{0.99}$. Then it is easy to calculate the following critical exponents from the cusp model for the fluid:

$$\begin{aligned} \alpha = \alpha' = 2 - 2\lambda = 0.02, & \quad \gamma = \gamma' = \lambda(4\theta - 2)/2\theta = \beta(\delta - 1) = 1.27, \\ \beta = \lambda/2\theta = 0.36, & \quad \delta = 4\theta - 1 = 4.56. \end{aligned} \tag{3.16}$$

From (3.5) one obtains

$$\nu = \nu_1 D_1/3 = 0.67. \tag{3.17}$$

For comparison we extract from Stanley (1971) and Ma (1976) the critical exponents shown in table 1.

Table 1. Critical exponents.

Material	α	α'	γ	γ'	β	δ	ν'
CO ₂ (St)	≈0.1	≈0.1	1.35	≈1.0	0.34	4.2	—
Xe (St)	—	<0.2	1.3	≈1.2	0.35	4.4	0.57
CO ₂ (Ma)	≈0.12	≈0.12	1.2	1.2	0.35	4.2	—
Xe (Ma)	0.08	0.08	1.2	1.2	0.34	4.4	—
He ³ (Ma)	<0.3	<0.2	1.15	1.15	0.36	—	—

4. Fractals

Finally, we have to explain what one may understand by a lattice with non-integral real dimension $D = 3.92$. To this end the notion of ‘dimension’ must be defined. Let M be a set and d a metric, $d : M \times M \rightarrow \mathbb{R}$ with $d(x, x) = 0 \forall x \in M$; $d(x, y) = d(y, x) > 0 \forall x, y \in M$ and $x \neq y$; $d(x, z) \geq d(x, y) + d(y, z) \forall x, y, z \in M$. (M, d) is a metric space. We define its Hausdorff–Besicovitch dimension in the following way (Mandelbrot 1977).

Let $U_\varepsilon(x) := \{y | d(x, y) \leq \varepsilon, \forall y \in M\}$ be the ball surrounding $x \in M$. We associate with $U_\varepsilon(x)$ a real positive number as an intrinsic Hausdorff measure for the volume of $U_\varepsilon(x)$, namely

$$0 < |U_\varepsilon(x)| = \varepsilon^D (\Gamma(\frac{1}{2}))^D / \Gamma(1 + \frac{1}{2}D) < \infty$$

where Γ is the Gamma function, and D is a real positive number to be specified later.

Let N be a compact subset of M . Then there exists a finite number $N(\varepsilon)$ of balls U_ε covering the whole subset N . The subset N has, therefore, the approximate finite measure

$$0 < |N| = N(\varepsilon) \varepsilon^D (\Gamma(\frac{1}{2}))^D / \Gamma(1 + \frac{1}{2}D) < \infty. \tag{4.1}$$

If, for every compact subset of M , the limit $\varepsilon \rightarrow 0$ of (4.1) is a unique, finite and non-zero number and independent of the way ε tends to zero, then M has Hausdorff–Besicovitch dimension D . This condition fixes the value of D , because the inequality $0 < \lim[N(\varepsilon) \varepsilon^D (\Gamma(\frac{1}{2}))^D / \Gamma(1 + \frac{1}{2}D)] < \infty$ could not hold for all $N \subseteq M$ if D were arbitrary.

In contrast to the topological definition of dimension, we are, of course, no longer confined to integer dimensions, $D \in \mathbb{N}_0 \equiv \{0, 1, 2, \dots\}$. Spaces with $D \notin \mathbb{N}_0$ have been introduced and are called by Mandelbrot (1977) ‘fractals’.

We illustrate this by a fractal object called the ‘Sierpinski sponge’ (figure 2). One cannot simply sketch this fractal, but only define it in an iterative geometric way. Suppose we are given a three-dimensional cube with sides of length one. If we divide each side into three we obtain 27 subcubes. We remove from the cube the central subcube and all those subcubes which have one face in common with the central subcube. There remain 20 subcubes. By this procedure the volume $V_0 = 1$ of our geometrical object is reduced by a factor $\frac{20}{27}$ and its surface area $F_0 = 6$ is enlarged by a factor $\frac{4}{3}$.

Repeating the above construction once for each of the remaining subcubes produces the same reduction of the volume and enlargement of the surface area for the remaining geometrical object. Thus, if the construction is iterated n times, the resulting object has volume

$$V = (\frac{20}{27})^n \tag{4.2}$$

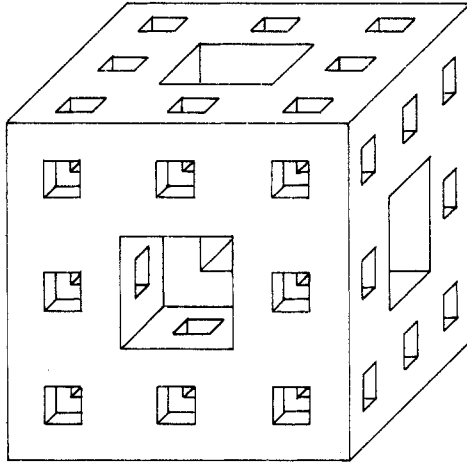


Figure 2. Construction of the Sierpinski sponge up to $n = 2$.

and surface area

$$F = 6\left(\frac{4}{3}\right)^n. \tag{4.3}$$

From (4.2) and (4.3) we see that $V \rightarrow 0$ and $F \rightarrow \infty$ as $n \rightarrow \infty$. If $n = \infty$ the object is called the ‘Sierpinski sponge’. This is a fractal whose three-dimensional volume is zero and whose two-dimensional surface area is infinitely large.

To determine the finite measure of this fractal, we use definition (4.1) with $\varepsilon = (\frac{1}{3})^n$ and $D = \lg(20)/\lg(3) \approx 2.7268$. Then $N(\varepsilon) = 20^n$, and by virtue of the identity $x = 10^{\lg(x)}$ one finds that

$$|N(\varepsilon)| = (\Gamma(\frac{1}{2})^D / \Gamma(1 + \frac{1}{2}D)) 20^n (\frac{1}{3})^{nD} = (\Gamma(\frac{1}{2})^D / \Gamma(1 + \frac{1}{2}D)) 10^{n \lg(20)} (10^{-\lg(3)})^{n \lg(20)/\lg(3)}$$

i.e. $0 < |N(\varepsilon)| = \Gamma(\frac{1}{2})^D / \Gamma(1 + \frac{1}{2}D) < \infty$. Since $N(\varepsilon)$ is actually independent of ε , we may choose $\varepsilon = 0$. Therefore,

$$N = N(0) = \Gamma(\frac{1}{2})^{\lg(20)/\lg(3)} / \Gamma(1 + \lg(20)/\lg(3)) \approx 0.51$$

is the measure of the fractal ‘Sierpinski sponge’.

Fractals have the following important properties.

(1) The definition (4.1) of the dimension of a fractal simplifies with $r = 1/\varepsilon$ to the following (Mandelbrot 1977):

$$D = \lim_{\varepsilon \rightarrow 0} \{ \lg[N(\varepsilon)] / \lg(1/\varepsilon) \} = \lim_{r \rightarrow \infty} \{ d \lg[N(r)] / d \lg(r) \}. \tag{4.4}$$

(2) Although a fractal seems to be a totally inhomogeneous object when seen from outside by embedding it in Euclidean space (e.g. figure 2), the fractal is actually a completely homogeneous medium (like a Euclidean space) when looked at within its own fractal dimension, i.e. if one is sitting in it. This is why the fractal possesses in a natural way the property of self-similarity, i.e. each part of it is similar to the whole object. Self-similarity is the reason for scaling laws, because from $N(r) \propto r^{-D}$ we obtain $N(ar) \propto a^{-D} N(r)$.

(3) Integration and differentiation, defined on spaces with $D \in \mathbb{N}$, can be extended to fractals (Mandelbrot 1977). Therefore equations like (2.7) remain true also for fractals, because $\int_L d^D z H(e(z), g)$ has a definite meaning.

(4) Suppose that there exists a minimum length scale ε_0 in the fractal such that all points x, y of the fractal satisfy $d(x, y) \geq \varepsilon_0$. Then we call the resulting object a fractal lattice, because for $D \in \mathbb{N}$ it reduces to a normal D -dimensional lattice.

There are some direct approaches to applying the concept of fractals to physical phenomena appearing in phase transitions (e.g. Stanley *et al* 1976, Stanley 1977, Mandelbrot 1977, Gefen *et al* 1980). The hierarchical clustering of molecules in a D -dimensional system is interpreted as formation of a fractal with *effective* dimension $D_p < D$. If we try to transcribe this picture to the abstract lattice, we suspect the appearance of clustering for the correlated regions of $e(z)$ in the fractal lattice with $D = 3.92$. The effective dimension of the clustered subset approaches $D_p = 3$ as $t \rightarrow 0$. Nevertheless, this conjecture is without any proof so far and not the aim of this paper. We confine ourselves to the existence of spaces with ‘broken dimension’ without physical interpretation.

Some further examples for the occurrence of fractals in physical sciences are found in information theory (Berger and Mandelbrot 1963) and are also encountered in the theory of electromagnetic waves (Berry 1979a, b).

5. Conclusions

In §§ 2 and 3 we constructed a model for a one-component fluid, representing it by a fractal lattice with dimension $D = 3.92$. The free energy per unit volume of the fractal lattice turned out to be diffeomorphic to an unfolding of the cusp singularity. The relation between the mathematical parameters x, a_1, a_2 of the cusp normal form for the free energy and the physical parameters ρ, t, p was established by applying scaling arguments in the lattice and in the fluid. This relation was non-analytic for $D = 3.92$.

However, in (3.13) we can choose $\eta = 0$ and then the concept of fractals is not necessary for critical and tricritical phenomena, but its introduction is imperative for tetracritical and higher-order phenomena (Güttinger and Keller 1981). If we choose $\eta = 0$ as in the Landau theory, then (3.13) reduces to $D/2 - 2 = 0$ or $D = 4$. For the butterfly singularity (describing tricritical phenomena), equation (3.11) remains unchanged and (3.12) becomes $x \propto s^{D/6}$. Consequently, we find $D = 3$ for the dimension of the lattice model describing a tricritical point. However, for the star singularity, which represents a tetracritical phenomenon, the lattice dimension should be $D = \frac{8}{3}$. This is in accordance with the fact that for each of the dimensions 4, 3, $\frac{8}{3}$ one observes the appearance of a new stable type of Hamiltonian for the fixed point in renormalisation group theory (Ma 1976, Sinai 1977, Amit 1978).

Table 2. Relation between lattice dimension, standard normal forms and order of phase transitions.

Phenomena	Lattice free energy	Lattice dimension ($\eta = 0$)
Critical	Cusp	4
Tricritical	Butterfly	3
Tetracritical	Star	$\frac{8}{3}$
...

The calculation of n -critical exponents for the butterfly ($n = 3$) and the star singularity ($n = 4$) within the framework of the above proposed lattice model will be discussed elsewhere (Güttinger and Keller 1981).

The calculated critical exponents agree well, though not exactly, with the experimental values. This seems to be due to the fact that some approximations have been made. First, we have required a highly *isotropic* lattice. Second, we have assumed the lattice to be *infinitely* extended. Third, we have required the possibility of formation of *unit volumes*. Finally, the restriction to *nearest-neighbour* interaction in a (3.92)-dimensional lattice may be relaxable. Despite all these approximations, most of our theoretical values lie within the range of experimental values shown in table 1.

Acknowledgment

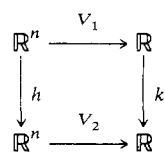
The author expresses his gratitude to Professor W Güttinger and Dr F J Wright for many helpful discussions.

Appendix

This section includes two topics for readers not familiar with the application of catastrophe theory to phase transitions. For a modern and detailed review we refer for example to Poston and Stewart (1978), Vendrik (1979) or Stewart (1980).

(a) *Brief exposition of Thom's theorem classifying structurally stable unfoldings*

Let \mathbb{R}^n denote n -dimensional real Euclidean space. A smooth potential function V is an infinitely differentiable mapping $V: \mathbb{R}^n \rightarrow \mathbb{R}$. Two potentials V_1 and V_2 are C^∞ equivalent ($V_1 \underset{C^\infty}{\sim} V_2$) if there exist diffeomorphisms $h: \mathbb{R}^n \rightarrow \mathbb{R}^n$ and $k: \mathbb{R} \rightarrow \mathbb{R}$ such that $V_2 \circ h = k \circ V_1$, i.e. if the diagram



is commutative. A diffeomorphism is a reversible C^∞ function (infinitely often differentiable and each time reversible).

Two potentials are near in the C^∞ topology if they and all their derivatives are close on each compact subset of \mathbb{R}^n (Whitney topology). A potential V is structurally stable if within some neighbourhood of V (in the C^∞ topology) all potentials are C^∞ equivalent. Structural stability is guaranteed by a non-vanishing Hessian, i.e. if for all $\mathbf{m} \in \mathbb{R}^n$

$$\bar{H}(\mathbf{m}) = \det \begin{bmatrix} \partial^2 V(\mathbf{m})/\partial x_1 \partial x_1 & \dots & \partial^2 V(\mathbf{m})/\partial x_1 \partial x_n \\ \vdots & & \vdots \\ \partial^2 V(\mathbf{m})/\partial x_n \partial x_1 & \dots & \partial^2 V(\mathbf{m})/\partial x_n \partial x_n \end{bmatrix} \neq 0$$

(x_1, \dots, x_n denote local coordinates in \mathbb{R}^n).

A smooth potential function $V_0(\mathbf{m})$ has a singularity at \mathbf{m}_0 if its derivative $\nabla V_0(\mathbf{m}_0)$ vanishes (∇ is the Nabla operator with respect to \mathbf{m}). If in addition $\bar{H}(\mathbf{m}_0) = 0$ then $V_0(\mathbf{m}_0)$ is not structurally stable; there exist potentials V near V_0 (in the C^∞ topology) which are not C^∞ equivalent to V_0 .

Neglecting further subtleties as 'germs', a k -parameter unfolding of V_0 is a smooth family $W(\mathbf{m}, \boldsymbol{\varepsilon})$ such that $W(\mathbf{m}, \mathbf{0}) = V_0(\mathbf{m})$ and $\boldsymbol{\varepsilon} \in \mathbb{R}^k$. V_0 is a singularity of the family $W(\mathbf{m}, \boldsymbol{\varepsilon})$. The number k is called the codimension of the unfolding.

A universal unfolding is one with minimal codimension and the property that all other unfoldings can be induced from a universal unfolding. Let the codimension be finite; then one can state that a universal unfolding of V_0 is given by the family

$$W(\mathbf{m}, \mathbf{v}) = V_0(\mathbf{m}) + \sum_{j=1}^k v_j g^j(\mathbf{m})$$

where k is as small as possible. A universal unfolding is unique up to equivalence and 'almost all' potentials V near V_0 are C^∞ equivalent to one member of the family W . 'Almost all' means that in the C^∞ topology (Whitney topology) the family W forms an open dense subset in the space of all potentials near V_0 .

Furthermore, this implies the structural stability of the family $W(\mathbf{x}, \mathbf{v})$, because any small perturbation of a member of W is equivalent to another member of W . The theorem of Thom classifies the structurally stable unfoldings.

Thom's theorem. Given a smooth family of potentials $W(\mathbf{x}, \mathbf{v})$; $W: \mathbb{R}^n \times \mathbb{R}^k \rightarrow \mathbb{R}$, then for $k \leq 4$ almost any singularity of W is equivalent to one of a small number of types called 'elementary catastrophes'. The number of types depends only on k and not on n .

This means that almost all structurally stable unfoldings are equivalent to some normal form, the normal form being defined as structurally stable unfolding of an elementary catastrophe.

For reasons of simplicity, we have formulated elementary catastrophe theory only for smooth potential functions on Euclidean spaces; nevertheless, the theory is true also for smooth potential functions on real manifolds.

In the application of catastrophe theory to thermodynamics, $E = \mathbb{R}^n$ represents a space of internal state variables (e.g. Schulman and Revzen 1972, Schulman 1973, Vendrik 1979) and $U = \mathbb{R}^k$ represents a space of external control variables. The potential $V: E \times U \rightarrow \mathbb{R}$ is a thermodynamic potential, usually considered as some kind of free energy (e.g. Schulman 1973) or the Gibbs potential (e.g. Dukek 1979).

(b) *The cusp normal form and van der Waals equation*

This topic illustrates one ingenious way of applying catastrophe theory to phase transitions. The example chosen is very famous amongst theoreticians engaged in catastrophe theory (e.g. Fowler 1972, Poston and Stewart 1978).

Consider the van der Waals equation of state for a one-component fluid:

$$(P + a/V^2)(V - b) = NkT. \quad (\text{A1})$$

Here P is the pressure, V is the volume and T is the absolute temperature of the fluid. The constants k and N are the Boltzmann constant and Avagadro's number respectively. The constants a and b depend on specific properties of the fluid's molecules.

According to Tisza's rule, the critical point $(P_c, V_c, T_c)^T$ is determined by solving (A1) and the additional equations

$$(\partial P/\partial V)_T = 0, \quad (\partial^2 P/\partial V^2)_T = 0.$$

This yields $P_c = a/27$, $V_c = 3b$ and $T_c = 8a/27Nkb$. Inserting the reduced coordinates

$$p = (P - P_c)/P_c, \quad v = (V - V_c)/V_c, \quad t = (T - T_c)/T_c,$$

in equation (A1) leads by some elementary operations to

$$v^3/(v + 1)^3 + \frac{1}{3}(8t + p)v/(v + 1) - \frac{1}{3}(8t - p) = 0. \tag{A2}$$

Introducing the 'order parameter' $x = -v/(v + 1)$, the above equation can be rewritten as

$$x^3 + \frac{1}{3}(8t + p)x - \frac{1}{3}(8t - p) = 0. \tag{A3}$$

The linear reversible coordinate transformation $B : (x, p, t)^T \rightarrow (y, a_1, a_2)^T$, specified by

$$B = \begin{bmatrix} 1 & 0 & 0 \\ 0 & \frac{1}{3} & -\frac{8}{3} \\ 0 & \frac{1}{3} & \frac{8}{3} \end{bmatrix}, \quad B^{-1} = \begin{bmatrix} 1 & 0 & 0 \\ 0 & \frac{3}{2} & \frac{3}{2} \\ 0 & -\frac{3}{16} & \frac{3}{16} \end{bmatrix},$$

transforms (A3) into

$$y^3 + a_2y + a_1 = 0. \tag{A4}$$

Integrating equation (A4) once (with respect to y) yields the real potential function

$$V(y, a_1, a_2) = \frac{1}{4}y^4 + \frac{1}{2}a_2y^2 + a_1y. \tag{A5}$$

This potential function is identical to the normal form of the unfolding of the cusp singularity $V_2(y, 0, 0) = \frac{1}{4}y^4$. (The expression 'normal form of the unfolding of the cusp singularity' is usually abbreviated by the terms 'cusp potential' or 'cusp normal form'.)

The cusp singularity has codimension two and is C^∞ equivalent to nearly all singularities with codimension two. A linear and reversible transformation like B is naturally a C^∞ function (all derivatives are vanishing), and likewise B^{-1} is a C^∞ function, i.e. B is a diffeomorphic transformation between the cusp singularity and some 'physical singularity' leading to the van der Waals equation of state.

Nevertheless, B diffeomorphic seems to be a chance hit, true for the special case of the van der Waals equation of state. If one constructs a mathematical description of a physical system governed by some real potential function, and classifies the possible singularities of that potential function according to Thom, no information about the relation between the variables of that potential and real physical quantities is necessary for the classification. The classification is determined solely by the codimension of the potential's singularity (Poston and Stewart 1978, Stewart 1980).

Let us return from this deviation to further discussion of the above example. The macroscopic states are—according to the Maxwell convention—given by the absolute minima of $V_2(y, a_1, a_2)$. A phase transition of the first kind occurs for competing minima, i.e. along the line $C = \{(a_1, a_2) | a_1 \equiv 0 \text{ and } a_2 < 0\}$ (coexistence line). A transformation with B^{-1} leads to the coexistence line in physical coordinates (phase diagram)

$$P = 4(P_c/T_c)(T - T_c) + P_c. \tag{A6}$$

For water steam this line coincides for small $(P - P_c)/P_c$ and $(T - T_c)/T_c$ with the coexistence line calculated from the van der Waals equation with the help of Maxwell's construction (Keller 1979).

Some straightforward calculations (e.g. Poston and Stewart 1978, Keller 1979), starting with (A5) and using the linearity of B^{-1} , result in the critical exponents $\beta = 0.5$, $\delta = 3$, $\gamma = 1$, $\gamma' = 1$, $\alpha = 0$, $\alpha' = 0$. These exponents disagree with the experimental results (e.g. Stanley 1971).

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